

Optical Attenuation in Liquid Argon

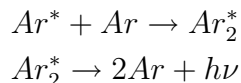
Alex Wright

March 3, 2011

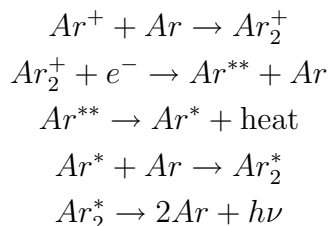
Abstract

One frequently heard explanation for the transparency of liquid argon to its own scintillation light, that the optical emission is due to a dimer which is not present in the normal liquid, is incorrect. A more correct explanation is given.

It is often argued that noble liquid detectors should be extremely transparent to their own scintillation light because the optical emission occurs from a neutral dimer excited state:



for direct excitations or



for ionizations [1] (in these equations * and ** denote excited electronic states, and + denotes an ionized atom/molecule). The frequency of the emitted radiation is therefore determined by the structure of the Ar_2 molecular orbitals, and to the extent that LAr is devoid of dimers, the attenuation lengths in LAr should be very long.

However, the binding energy of Ar_2 is 12.3 meV [2]. This is more than 100 times less than the binding energy of Ar_2^+ , but it is still significant compared to the thermal energy ($kT(89\text{ K})=5.9\text{ meV}$); this is unsurprising given that both liquification and dimerization occur because of the same Van der Waal's forces. It is difficult to estimate the entropy of dimerization in a liquid in order to allow a proper calculation of the dimer-monomer equilibrium constant, but a zeroth order estimate of the partition fraction using the Boltzmann distribution gives $e^{-2.08} = 0.12$, meaning that 88% of argon atom pairs should be in the lower (dimer) state. This is almost certainly an overestimate, but the conclusion that a reasonable fraction of the atoms in liquid argon are in the dimer state is probably a good one.

The cross section for the re-absorption of a photon by its emitting atom at wavelength λ can be estimated as

$$\sigma(\nu) = \frac{\lambda^2}{8\pi t_{sp}} g(\nu) \quad (1)$$

where t_{sp} is the spontaneous emission lifetime of the state, and $g(\nu)$ is the line shape of the corresponding emission [3]. For an average effect, we assume a Lorentian $g(\nu)$ with a FWHM of $\Delta\nu$, which gives

$$\sigma_o = \frac{\lambda^2}{4\pi^2 t_{sp} \Delta(\nu)}. \quad (2)$$

For liquid argon singlet state emission (which should dominate the re-absorption), we have $\lambda = 126.8\text{ nm}$ and $\Delta\nu \sim 10\text{ nm}$, giving $\sigma_o = 3.6 \times 10^{-18}\text{ cm}^2$. For a liquid argon density of $1.4\text{ g/cm}^3 = 2.1 \times 10^{22}\text{ atoms/cm}^3$, and assuming 100% dimerization, this gives a tiny attenuation length, of order $1 \times 10^{-5}\text{ m}$. This is quite short, and in conflict with the 60-90 cm attenuation lengths (dominated by Rayleigh scattering) and $>1\text{ m}$ absorption lengths that are experimentally observed [4, 5]

The functionality of our experiment is saved by a massive Stokes' shift which is caused by the binding energy being significantly stronger, and hence the equilibrium internuclear distance significantly shorter, in Ar_2^* than in Ar_2 . Thus, the molecular transition from the excited state to the ground state results in a vibrationally excited form of the (electronic) ground state, which subsequently relaxes to its equilibrium position (in fact the excited dimer is unbound, so it will break apart and subsequently reform). This energy structure is shown in Figure 1. In order to re-absorb the emitted photon, a dimer would have to be in a vibrational excited state almost 1 eV above ground state (again, such a state would actually be unbound so this "state"

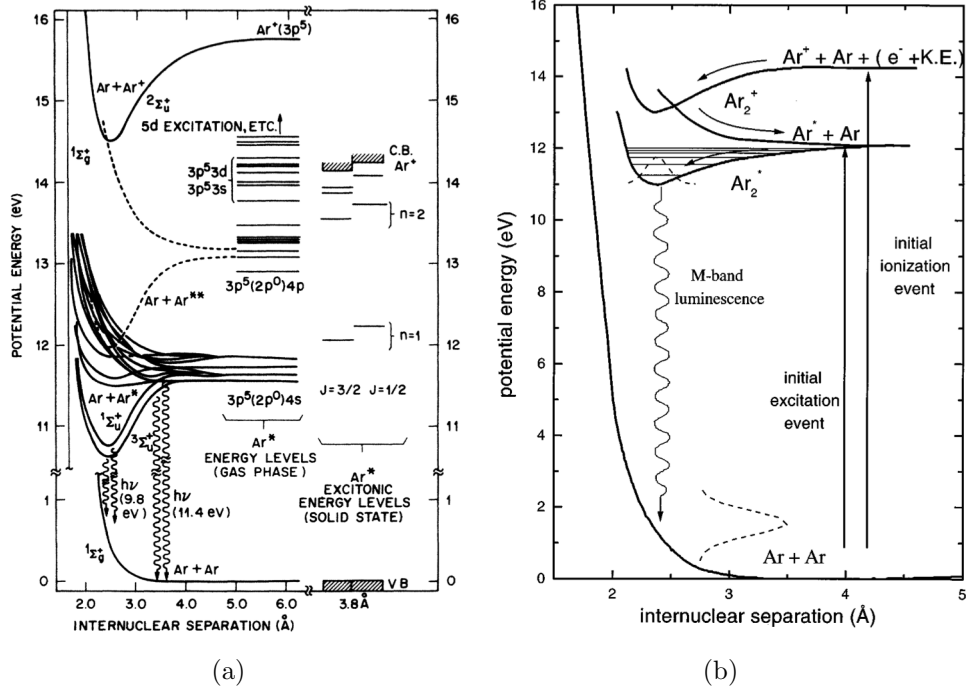


Figure 1: (a) The potential energy diagram for gas phase argon atoms as a function of internuclear distance [6]. The 9.8 eV transition dominates the emission spectrum; the short-lived singlet and long-lived triplet states can be seen in the diagram. (b) A simplified version of the diagram, this time for solid argon, showing schematically the path by which ionization-type excitations are thought to cause photon emissions [7]. The very shallow potential well of the ground state neutral dimer can be seen at the bottom of the Figure.

would be formed only during collisions between atoms). Roughly estimating the “population” of this “excited state” using the Boltzmann distribution again gives $e^{-1.0/0.006} \sim 10^{-73}$, so re-absorption of the scintillation light is indeed negligible. I note that this effect is also the operational basis for excimer lasers.

References

- [1] T. Doke et. al., “Estimation of Absolute Photon Light Yields in Liquid Argon and Xenon for Relativistic (1 MeV) Electrons.” Nucl. Inst. Meth. A **291** (1989) 617-620.

- [2] T. Pradeep et al., “Photoelectron Spectroscopy of Rare Gas Dimers Revisited: Vibrationally Resolved Photoelectron Spectrum of Argon Dimer.” J. Chem. Phys. **98** (1993) 5269-5275.
- [3] B.E.A. Saleh and M.C. Teich. *Fundamentals of Photonics*. John Wiley & Sons, Inc., New York, 1991. pp 442-443.
- [4] N. Ishida et al., “Attenuation Length Measurements of Scintillation Light in Liquid Rare Gases and Their Mixtures Using an Improved Reflection Suppressor.” Nucl. Inst. Meth. A **384** (1997) 380-386.
- [5] Icarus, personal communication with Luca.
- [6] C.T. Reimann et al., “Electronically Stimulated Sputtering and Luminescence from Solid Argon.” Phys. Rev. B **37** (1988) 1455-1473.
- [7] D.E. Grosjean et al., “Absolute Luminescence Efficiency of Ion-Bombarded Solid Argon.” Phys. Rev. B **56** (1997) 6975-6981.